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Charged excitons in ZnSe-based QWs

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Abstract. We report on magneto-optical studies of ZnSe/(Zn,Mg)(S,Se) and ZnSe/(Zn,Be,Mg)Se quantum wells with n-type and p-type modulation doping. Negatively and positively charged excitons related to the heavy-hole exciton state are found and identified by their polarization properties. Exciton and trion parameters (radiative and nonradiative dampings, g factors) are determined.

The existence of charged exciton complexes (trions) in semiconductors has been predicted by Lampert in 1958 [1]. In analogue with charged hydrogen ions H^- and H_2^+ the negatively charged exciton (X^-), consisting of two electrons and one hole, and the positively charged exciton (X^+), formed by two holes and one electron, have been suggested. However, the first clear experimental proof of the X^- existence has appeared quite recently in 1993. Kheng *et al.* reported the observation of the negatively charged exciton state in a CdTe-based quantum well (QW) structure with a two-dimensional electron gas (2DEG) of low density [2]. It follows from the calculation of the charged exciton binding energy that this complex is only weakly bound in bulk materials (about 5% of exciton Rydberg energy), which hinders its experimental observation in 3D systems. However, reduction of dimensionality of the system down to quasi-two-dimensional one strongly favors the trion stability and increases its binding energy up to 20–45% of the Rydberg energy. In recent years negatively charged excitons have been studied intensively in CdTe- and GaAs-based QWs [3, 4]. The observation of positively charged excitons has also been reported for these structures [5–7].

Being scaled with the exciton Rydberg the trion binding energy can be enhanced considerably by the proper choice of the material system. The logical step after GaAs (with exciton binding energy of 4.2 meV) and CdTe (10 meV) is to investigate ZnSe-based structures (20 meV) in order to improve the Coulomb interaction. The technique of molecular-beam epitaxy (MBE) is well developed for the growth of ZnSe-based QW structures. However, the mostly studied structures contain (Zn,Cd)Se ternary alloy QWs barriered by ZnSe layers. In these structures the exciton resonances are broadened significantly due to alloy fluctuations, which makes highly-resolved optical spectroscopy of exciton and trion states very difficult. We are aware of one report on X^- state observation in (Zn,Cd)Se/ZnSe QW only [8]. Very recently the growth of high-quality lattice-matched ZnSe/(Zn,Mg)(S,Se) QWs with exciton inhomogeneous broadening less than 1 meV has been published [9, 10]. In this paper we report on magneto-optical studies of negatively and positively charged excitons in ZnSe/(Zn,Mg)(S,Se) and ZnSe/(Zn,Be,Mg)Se QWs. Trion states related to both heavy-hole and light-hole excitons are found and their parameters are examined as a function of QW width and in external magnetic fields.

A set of $ZnSe/Zn_{0.86}Be_{0.06}Mg_{0.08}Se$ single QW (SQW) structures with QW width varied from 30 Å up to 200 Å was grown by MBE on (100) GaAs substrates. The ZnSe QWs were confined by 1000 Å thick barriers. A total band gap discontinuity between QW

and barrier materials of 230 meV is distributed in ratio 70/30 between the conduction and valence bands. The structures were nominally undoped. $ZnSe/Zn_{0.89}Mg_{0.11}S_{0.18}Se_{0.82}$ SQWs were also grown by MBE on (100) GaAs substrates. ZnSe SQWs were located between $Zn_{0.89}Mg_{0.11}S_{0.18}Se_{0.82}$ barriers of 1000 Å- and 500 Å thickness. The total band gap discontinuity of 200 meV is distributed about equally between the conduction and valence band edges. Results for three structures are reported here. The first one is nominally undoped with a residual concentration of the 2DEG in the 100 Å SQW $n_e \leq 10^{10}\,\mathrm{cm}^{-2}$ due to the weak n-type background conductivity of the barriers. The second structure has $n_e = 9 \times 10^{10}\,\mathrm{cm}^{-2}$ provided by a modulation doped layer (30 Å thick, Cl doped to a level of $5 \times 10^{17}\,\mathrm{cm}^{-3}$) separated by a 100 Å spacer from a 100 Å SQW. The third sample was p-type doped with nitrogen through the whole thickness of the barrier layers excluding an 120 Å SQW and 30 Å spacers on both sides of the SQW. The carrier concentration of the two-dimensional hole gas (2DHG) is about $n_h \approx 3 \times 10^{10}\,\mathrm{cm}^{-2}$. Photoluminescence (PL), reflectivity and spin-flip Raman scattering (SFRS) spectra were measured at 1.6 K and in magnetic fields up to 7.5 T applied perpendicular to the QW plane (Faraday geometry). Circularly polarized light was analyzed by means of achromatic quarter-wave plates. The signal was dispersed by a 1-m spectrometer and detected by either a charge-coupled device (CCD) or by a photomultiplier.

In Fig. 1(a,b) PL and reflectivity spectra of 30 Å- and 200 Å thick QWs are plotted. In low-temperature emission spectra two lines corresponding to the heavy-hole exciton (X) and X^- are clearly seen (detailed identification of the optical resonances by means of magneto-optics in fields up to 20 T was reported in Ref. [10]). For the 200 Å the X^-

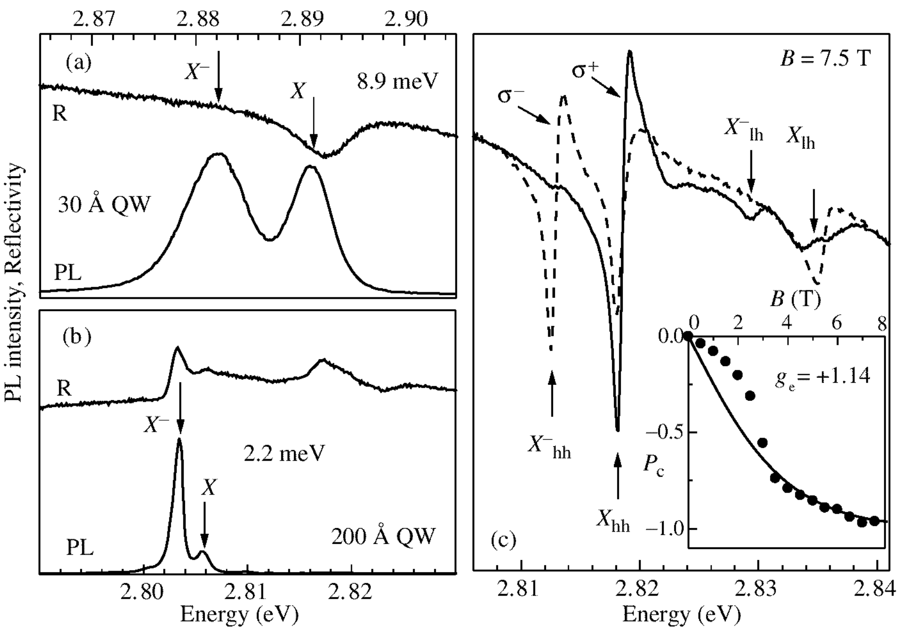

Fig. 1. Photoluminescence and reflectivity spectra of $ZnSe/Zn_{0.86}Be_{0.06}Mg_{0.08}Se$ SQWs with well width of 30 Å (a) and 200 Å (b) taken at 1.6 K and at a zero magnetic field. In the panel (c) reflectivity spectra of a 100 Å thick $ZnSe/Zn_{0.89}Mg_{0.11}S_{0.18}Se_{0.82}$ SQW with $n_e \approx 9 \times 10^{10}\,\mathrm{cm}^{-2}$ detected in two circular polarizations are presented. An inset shows the polarization degree of X^-_{hh} transition: experimental data (circles) and calculated dependence (solid line).

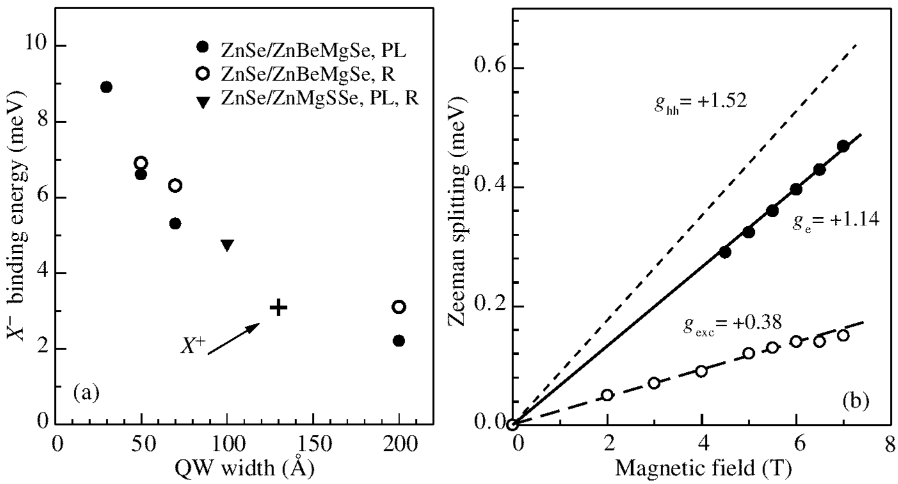

Fig. 2. (a) Trion binding energies as a function of QW width for the studied ZnSe-based structures. Data points evaluated from photoluminescence (PL) and reflectivity (R) spectra are shown. (b) Zeeman splittings of heavy-hole excitons and carriers in a 100 Å thick $ZnSe/Zn_{0.89}Mg_{0.11}S_{0.18}Se_{0.82}$ SQW. Experimetal data are shown by circles and linear interpolations are represented by lines.

line is also intensive in the reflectivity spectrum, however, it is not detectable for the 30 Å QW. The main reason for that is a difference in 2DEG concentrations (we estimate from the optical spectra $n_e \geq 10^{11}$ cm^{-2} for the 200 Å QW and $n_e \leq 10^{10}$ cm^{-2} for the 30 Å QW), which in these nominally undoped structures is supplied by shallow donors of the barrier layers. In narrow QWs the electron quantum-confined energy level is higher than the barrier donor levels and electron collection into the QW is reduced significantly.

The binding energy of the X^- state defined as the energy difference between X^- and X is plotted in Fig. 2(a) as a function of QW width. It increases by a factor of 4 with well width reduction from 200 Å down to 30 Å and amounts to 9 meV. For the 100 Å QW the X^- binding energy is 17% of the exciton binding energy in the QW, which value of 30 meV was determined from the magneto-exciton fan chart [10].

In Fig. 2(b) experimental results on the Zeeman splitting of the exciton and carriers in the 100 Å $ZnSe/Zn_{0.89}Mg_{0.11}S_{0.18}Se_{0.82}$ QW are collected. The exciton splitting with a g factor $g_{exc} = +0.38$ was deduced from the reflectivity spectra and that of the electron with $g_e = +1.14$ was measured by means of SFRS. The heavy-hole g factor was calculated by means of the equation $g_{hh} = g_{exc} + g_e = +1.52$ [11].

The strong polarization of the X^- absorption in magnetic fields is a fingerprint of the trion which allows to distinguish clearly trion resonance from the excitonic one [2]. This is due to the singlet spin structure of the trion ground state, i.e. spins of two electrons involved in the X^- complex should be oriented antiparallel. As a result, when a 2DEG is totally polarized by the magnetic field X^- can be excited optically only for one circular polarization of light (namely the σ^- polarization in ZnSe-based QWs with a positive electron g factor $g_e = +1.14$ measured by SFRS (see Fig. 2(b)). At 7.5 T the X^-_{hh} transition in the reflectivity spectra is totally polarized (see Fig. 1(c)). Its oscillator strength became twice stronger in σ^- polarization, comparing with the zero field value, and vanishes for σ^+ polarization keeping the integral oscillator strength constant.

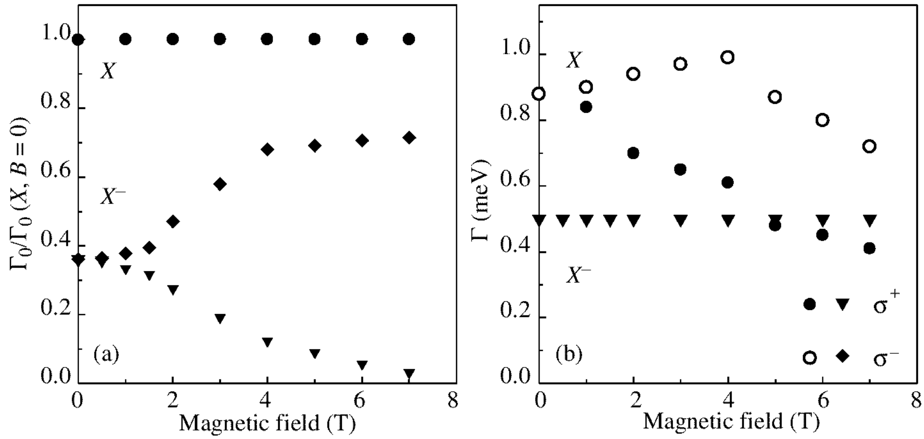

Fig. 3. Magnetic-field dependences of radiative (Γ_0) and nonradiative (Γ) dampings for exciton and trion transitions in a 100 Å thick $ZnSe/Zn_{0.89}Mg_{0.11}S_{0.18}Se_{0.82}$ SQW with $n_e \approx 9 \times 10^{10}\,\text{cm}^{-2}$. Results for two circular polarized components of reflectivity spectra are presented at $T = 1.6$ K.

A procedure described in Ref. [12] was used to fit the reflectivity spectra and to deduce the excitonic parameters like radiative damping Γ_0, which corresponds to the exciton oscillator strength, and the nonradiative damping Γ, which in the studied structures is dominated by inhomogeneous broadening. The parameters determined as a function of magnetic fields are presented in Fig. 3. We stress here that Γ_0 for the exciton and Γ for the trion do not vary in magnetic fields. The difference in Γ for the exciton components with opposite circular polarizations is due to an effect of spin-dependent scattering of excitons with 2D electrons [13].

The degree of polarization of the trion transition calculated as $P_c = (\Gamma_0^+ - \Gamma_0^-)/(\Gamma_0^+ + \Gamma_0^-)$ is shown in the inset of Fig. 1(c) by circles. The equilibrium polarization of a nondegenerate 2DEG, calculated with the Boltzmann distribution, $g_e = +1.14$ and $T = 1.6$ K, is traced by a solid line. Experimental points coincide well with the line at fields above 3.7 T. Deviation from the Boltzmann distribution in low magnetic fields takes place for filling factors $\nu > 1$ [14]. In this case the Fermi-Dirac statistic describes the polarization properties of the 2DEG. We conclude from the inset of Fig. 1(c) that the condition $\nu = 1$ is achieved at a magnetic field of 3.7 T. This, in turn, allows to determine the concentration of the 2DEG $n_e = \nu eB/hc = 9 \times 10^{10}\,\text{cm}^{-2}$ (for details see [15]).

The X^-_{lh} transition, related to a negatively charged exciton associated with the light-hole exciton states, is observed at 7.5 T as a clearly resolved resonance 4.4 meV below the energy of the light-hole exciton (Fig. 1(c)). It is polarized contrary to the X^-hh states, which is in agreement with optical selection rules. The binding energy of X^-_{lh} is about 20% smaller than that of X^-_{hh} which is 5.5 meV at 7.5 T. To the best of our knowledge no detailed investigation of X^-_{lh} states has been reported so far. Trions associated with the light-hole exciton were observed in PL excitation spectra of GaAs/(Al,Ga)As QWs [3] and in the reflectivity spectra of monomolecular CdTe islands [16]. In both cases the X^-_{lh} binding energy is very close to that of X^-_{hh}.

A shake-up line related to X^-_{hh} has been detected in the emission spectra of the structure with $n_e \approx 10^{10}\,\text{cm}^{-2}$ [17] (see Fig. 4(a,b)). It moves towards lower energies with increasing

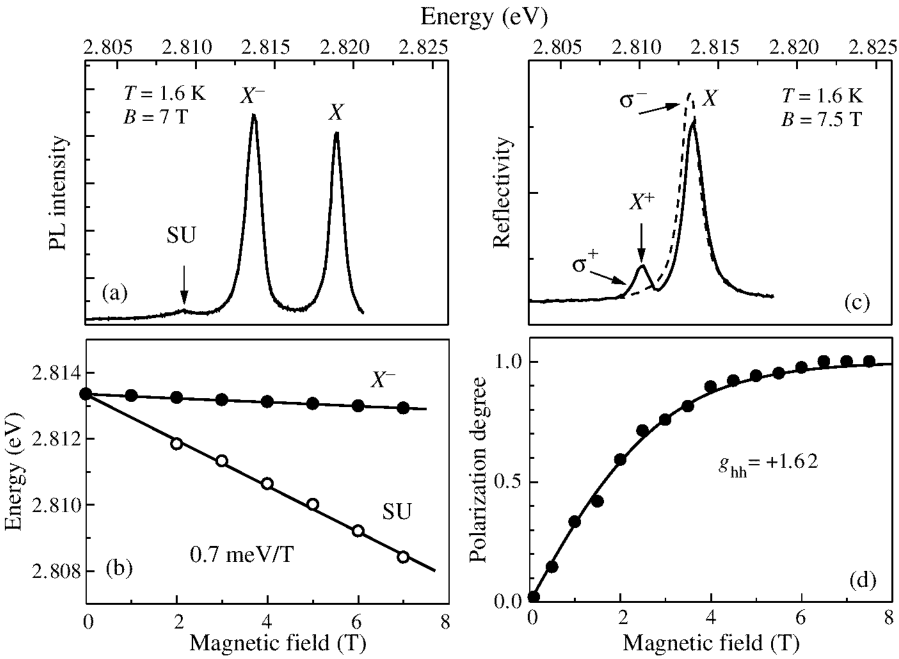

Fig. 4. A shake-up (SU) process in PL spectrum of a 100 Å thick $ZnSe/Zn_{0.89}Mg_{0.11}S_{0.18}Se_{0.82}$ SQW with $n_e \approx 10^{10}\,cm^{-2}$ (a) and magnetic-field dependence of the SU line (b). A positively charged exciton in reflectivity spectra of a 130 Å $ZnSe/Zn_{0.89}Mg_{0.11}S_{0.18}Se_{0.82}$ QW with $n_h \approx 3 \times 10^{10}\,cm^{-2}$ (c) and polarization dependence of X^+ line in magnetic fields: experiment (circles) and calculation for $T = 1.6$ K (line).

magnetic field with a slope of 0.7 meV/T, which is very close to the electron cyclotron energy of 0.725 meV/T (electron effective mass in ZnSe is $m_e = 0.16m_0$). This combined process is based on the recombination of an trion and a simultaneous excitation of a 2D electron from the zeroth to the upper Landau levels.

In Fig. 4(c) reflectivity spectra for the 130 Å thick SQW with a low-dense 2DHG with $n_h \approx 3 \times 10^{10}\,cm^{-2}$ are shown. A line of the positively charged exciton with a binding energy of 3.1 meV is observed in the spectrum. Similar to X^-_{hh} the X^+_{hh} line is totally polarized in magnetic fields above 6 T, but the sign of polarization is opposite. The magnetic-field dependence of the X^+_{hh} polarization degree (see circles in the inset of Fig. 4(d)) coincides remarkably well with the Boltzmann distribution calculated with the heavy-hole g factor $g_{hh} = +1.62$. This g factor value was deduced from the measured $g_{exc} = +0.48$ and $g_e = +1.14$. The value of the X^+_{hh} binding energy of 3.1 meV is close to that for the X^-_{hh} (see Fig. 2(a)), which is in agreement with the results reported for QW structures based on GaAs [6, 7], and CdTe [5].

To conclude, charged excitons shows up brightly in the optical spectra of ZnSe-based QWs. Small inhomogeneous broadening of excitonic transitions combined with a strong Coulomb interaction (5 times stronger than in GaAs-based QWs) allows us to suggest these structures as model objects for the investigation of exciton-electron interaction phenomena in semiconductors.

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